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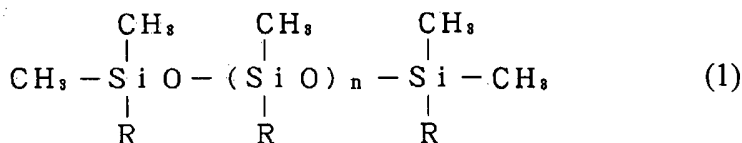
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(54) Title: WAXY ORGANOPOLYSILOXANE



(57) Abstract: A waxy organopolysiloxane, which has a chemical structure represented by below-given general formula (1), a melting point of 55 °C to 115 °C, a viscosity at 120 °C of 50 mPa•s or below, and a penetration degree at 50 °C of 15 or below:

$\text{R}(\text{CH}_3)_2\text{SiO}\{\text{R}(\text{CH}_3)\text{SiO}\}_n\text{SiR}(\text{CH}_3)_2$, is characterized by good flowability in a molten state, does not migrate in a compounded product at room temperature, but exudes at a sufficient degree when the compounded product is heated, and demonstrates a mold-release effect.

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DESCRIPTION**WAXY ORGANOPOLYSILOXANE****Technical Field**

[0001] The present invention relates to a waxy organopolysiloxane, in particular, to a
5 waxy organopolysiloxane that is not tacky at room temperature, has an appropriate melting
point, possesses good flowability in a molten state, does not migrate at room temperature
in a compounded product but exudes from the product at a sufficient rate when the product
is heated, and demonstrates good mold-release properties.

Background Art

10 [0002] It is known that an organopolysiloxane modified by long-chain alkyl groups
having on average more than 16 carbons is suitable for use as a wax in thermo-sensitive
transfer ribbons, thermo-sensitive paper, toners, etc.

[0003] It is desired that the aforementioned wax not be tacky, can sharply melt at a
predetermined temperature, and maintain certain hardness prior to melting. In other
15 words, it is desired that this wax has a narrow melting temperature range.

[0004] An example of such a product is a waxy organopolysiloxane having specific
melting characteristics and prepared by purifying a precursor with long-chain alkyl groups
(see Japanese Patent Application Laid-Open No. 2003-221445).

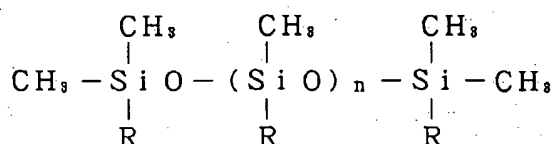
[0005] However, the waxy organopolysiloxane disclosed in the Japanese Patent
20 Application has insufficient flowability in a molten state and insufficient exudation from a
compounded product.

[0006] It is an object of the present invention to provide a waxy organopolysiloxane
that is not tacky at room temperature, has an appropriate melting point, is characterized by
good flowability in a molten state, does not migrate in a compounded product at room
25 temperature, but exudes at a sufficient rate when the product is heated, and demonstrates a
mold-release effect.

[0007] As a result of a study aimed at the accomplishment of the above object, the
inventors of the present patent application have found a waxy organopolysiloxane that has
a specific chemical structure and physical properties and solves all the problems of the
30 prior art. Thus the inventors arrived at the present invention.

Disclosure of Invention

[0008] A waxy organopolysiloxane of the invention comprises a modified organopolysiloxane of the below-given general formula (1) that has a melting point of 55°C to 115°C, a viscosity at 120°C of 50 mPa·s or below, and a penetration degree at 50°C of 15 or below:



[where R, independently, is:

- (a) a long-chain alkyl group with 20 to 100 carbon atoms or a long-chain organic group that contains heteroatoms;
 - (b) a hydrogen atom, an alkyl group with 2 to 19 carbon atoms, an aryl group with 6 to 10 carbon atoms, an aralkyl group with 7 to 14 carbon atoms, an alkoxy group with 1 to 8 carbon atoms, or an organic group with 2 to 19 carbon atoms that contains heteroatoms; or
 - (c) a methyl group;
- “n” is an integer within the range of 0 to 1000;
among the groups designated by R, the groups of Item (a) being contained in one molecule in the quantity of one or more.]

Effects of Invention

[0009] The waxy organopolysiloxane of the invention produces the following effects:

- (1) It is not tacky and has certain hardness at room temperature.
- (2) It has an appropriate melting point.
- (3) It is characterized by good flowability in a molten state.
- (4) At room temperature, it does not migrate (bleed/bloom) in a compounded product that contains this organopolysiloxane.
- (5) A compounded product that contains the waxy organopolysiloxane of the invention is not tacky at room temperature.
- (6) Heating of a product that contains the waxy organopolysiloxane of the invention to a predetermined temperature causes exudation of the waxy organopolysiloxane to the surface of the product. This phenomenon provides good mold-release properties.

Brief Description of the Drawings

- [0010] Figure 1 contains a GC chart obtained by measuring SiH1.
[0011] Figure 2 contains a GC chart obtained by measuring SiH2.
[0012] Figure 3 contains a GC chart obtained by measuring SiH3.
5 [0013] Figure 4 contains a GC chart obtained by measuring SiH4.
[0014] Figure 5 contains a GC chart obtained by measuring SiH5.
[0015] Figure 6 contains a GPC chart obtained by measuring SiH6.
[0016] Figure 7 contains a GC chart obtained by measuring SiH7.
[0017] Figure 8 contains a GC chart obtained by measuring SiH8.
10 [0018] Figure 9 contains a GC chart obtained by measuring SiH9.

Detailed Description of the Invention

[0019] The invention will be further described in more detail.

<Chemical Structure of the Waxy Organopolysiloxane>

15 [0020] The chemical structure of the waxy organopolysiloxane of the invention is shown by aforementioned general formula (1).

[0021] In aforementioned general formula (1), R independently designates (a) a long-chain alkyl group with 20 to 100 carbon atoms or a long-chain organic group that contains heteroatoms; (b) a hydrogen atom, an alkyl group with 2 to 19 carbon atoms, an aryl group with 6 to 10 carbon atoms, an aralkyl group with 7 to 14 carbon atoms, an alkoxy group with 1 to 8 carbon atoms, or an organic group with 2 to 19 carbon atoms that contains heteroatoms; or (c) a methyl group.

20 [0022] (a) The long-chain alkyl group with 20 to 100 carbon atoms or a long-chain organic group that contains heteroatoms, as a part of definition of R, may have a linear or branched structure. In the subsequent description, both the long-chain alkyl group and the long-chain organic group that contains heteroatoms will be referred to simply as "long-chain organic groups".

30 [0023] Normally, long-chain organic groups of Item (a) should have 20 to 100 carbon atoms, preferably 20 to 90 carbon atoms, more preferably, 20 to 80 carbon atoms, and still more preferably, 25 to 60 carbon atoms. If the number of carbon atoms in the long-chain organic groups is less than 20, the organopolysiloxane becomes oily rather than waxy. If, on the other hand, the number of carbon atoms exceeds 100, the organopolysiloxane either

acquires an extremely high melting point, or becomes too viscous in a molten state, and cannot be applied in wax with high flowability.

[0024] The long-chain organic groups may be represented by linear-chain alkyl groups $[\text{CH}_3(\text{CH}_2)_p-]$ (where "p" is an integer within the range of 19 to 99, preferably, within the range of 19 to 89, and more preferably, within the range of 19 to 79.) and branched alkyl groups. The linear alkyl groups are preferable. Examples of such long-chain alkyl groups are the following: $\text{CH}_3(\text{CH}_2)_{23}-$, $\text{CH}_3(\text{CH}_2)_{45}-$, etc.

[0025] Heteroatoms contained in the long-chain organic groups would be other than carbon atoms. Furthermore, heteroatoms of different types may be used simultaneously irrespective of their positions in the molecule. The long-chain organic groups with heteroatoms may be connected to the organopolysiloxane via alkylene groups with 2 to 15 carbon atoms or via alkylene with 2 to 15 carbon atoms -CO- groups. The total number of heteroatoms in the long-chain group may be 1 to 10, preferably 1 to 5, and even more preferably, 1 to 3. It is preferable to select heteroatoms from oxygen or nitrogen.

[0026] The following are examples of long-chain organic groups that contain heteroatoms: alkoxy carbonyl alkyl groups $[\text{CH}_3(\text{CH}_2)_p\text{O}(\text{CO})(\text{CH}_2)_q-]$; alkyl aminocarbonyl alkyl groups $[\text{CH}_3(\text{CH}_2)_p\text{NHCO}(\text{CH}_2)_q-]$; alkyl carbonyloxy alkyl groups $[\text{CH}_3(\text{CH}_2)_p\text{COO}(\text{CH}_2)_q-]$; alkoxy alkyl groups $[\text{CH}_3(\text{CH}_2)_p\text{O}(\text{CH}_2)_q-]$; alkyl carbonyloxy (2-hydroxy) propyloxy alkyl groups $[\text{CH}_3(\text{CH}_2)_p\text{COOCH}_2\text{CH}(\text{OH})\text{CH}_2\text{O}(\text{CH}_2)_q-]$; alkyl carbonylamino alkyl groups $[\text{CH}_3(\text{CH}_2)_p\text{CONH}(\text{CH}_2)_q-]$; alkyl aminocarbonylamino alkyl groups $[\text{CH}_3(\text{CH}_2)_p\text{NHCONH}(\text{CH}_2)_q-]$; alkoxy carbonylamino alkyl groups $[\text{CH}_3(\text{CH}_2)_p\text{O}(\text{CO})\text{NH}(\text{CH}_2)_q-]$; alkoxy carbonyl amino alkylamino alkyl groups $[\text{CH}_3(\text{CH}_2)_p\text{O}(\text{CO})(\text{CH}_2)_q\text{NH}(\text{CH}_2)_r-]$; and alkyl carbonylamino alkylamino alkyl groups $[\text{CH}_3(\text{CH}_2)_p\text{CONH}(\text{CH}_2)_q\text{NH}(\text{CH}_2)_r-]$; the number of carbon atoms being the same as specified earlier.

[0027] In the above formulae, "p" is an integer that provides the number of carbon atoms in the long-chain organic group within the range of 20 to 100, "q" is an integer of 1 to 15, preferably of 2 to 13, and "r" is an integer of 1 to 15, preferably of 2 to 10, with the proviso that "q + r" is an integer of 3 to 16.

[0028] In order to obtain the organopolysiloxane of the invention in a waxy state, a molecule should contain at least one of the groups of aforementioned Item (a).

[0029] Among the total $(n + 2)$ of groups designated by R, the proportion of Item (a) groups is normally within the range of 10 to 80%, preferably 25 to 75%.

[0030] The groups of Item (b), as a part of the groups designated by R, can be represented by hydrogen atoms, alkyl groups with 2 to 19 carbon atoms, aryl groups with 6 to 10 carbon atoms, aralkyl groups with 7 to 14 carbon atoms, alkoxy groups with 1 to 8 carbon atoms, or organic group with 2 to 19 carbon atoms that contain heteroatoms.

[0031] The alkyl groups with 2 to 19 carbon atoms can be exemplified by ethyl groups, propyl groups, isopropyl groups, butyl groups, isobutyl groups, hexyl groups, and octyl groups.

[0032] The aryl groups with 6 to 10 may be represented by phenyl and naphthyl groups, of which phenyl groups are preferable.

[0033] The aralkyl groups with 7 to 14 carbon atoms can be represented by benzyl and phenethyl groups.

[0034] Groups of Item (b) are by-products resulting from introduction of long-chain organic groups into the organopolysiloxane. In case of organic groups with 2 to 19 carbon atoms that contain heteroatoms, the aforementioned by-product groups comprise intermediate reactive groups formed in a chain-elongation reaction, hydrosilyl groups, hydroxyl groups, epoxy groups, or similar reactive groups, as well as groups resulting from capping and deactivating the reactive groups.

[0035] The aforementioned groups of Item (b) can be represented by hydroxyalkyl groups $[\text{HO}(\text{CH}_2)_q -]$, alkoxyalkyl groups $[\text{CH}_3(\text{CH}_2)_s\text{O}(\text{CH}_2)_q -]$; epoxypropyloxyalkyl groups $[\text{CH}_2(\text{O})\text{CHCH}_2\text{O}(\text{CH}_2)_q -]$; alkoxy (2-hydroxy) propyloxyalkyl groups $[\text{CH}_3(\text{CH}_2)_s\text{OCH}_2\text{CH}(\text{OH})\text{CH}_2\text{O}(\text{CH}_2)_q -]$; alkoxycarbonylalkyl groups $[\text{CH}_3(\text{CH}_2)_s\text{O}(\text{CO})(\text{CH}_2)_q -]$; and alkylcarbonyloxyalkyl groups $[\text{CH}_3(\text{CH}_2)_s\text{COO}(\text{CH}_2)_q -]$.

In these formulae, "s" is an integer of 0 to 13, preferably, 0 to 2; and "q" is the same as defined above. In the above groups, the total number of carbon atoms may be within the range of 2 to 19.

[0036] Among the total $(n + 2)$ of groups designated by R, the proportion of Item (b) groups is normally within the range of 0 to 50%, preferably 0 to 20%, and even more preferably, 0 to 5%.

[0037] Furthermore, the total proportion in terms of mass of Item (a) groups and Item (b) groups in the waxy organopolysiloxane of aforementioned general formula (1) normally should not exceed 95%, preferably 90%, and more preferably 85%.

[0038] If the total mass proportion of Items (a) and (b) groups in the organopolysiloxane exceeds 95%, it will be difficult to form a coating layer with surface characteristics inherent in a silicone wax (such as luster, smoothness, mold-release properties, etc.).

5 [0039] The groups of Item (c), as a part of the groups designated by R, are methyl groups. Among the total (n + 2) of groups designated by R, the proportion of methyl groups should normally be within the range of 20 to 90%, preferably, 25 to 75%. If in the organopolysiloxane the proportion of methyl groups among those designated by R is less than 20%, it will be difficult to form a coating layer with surface characteristics inherent in
10 a silicone wax (such as luster, smoothness, mold-release properties, etc.). If, on the other hand, the proportion of the methyl groups exceeds 90%, it would be difficult to obtain a waxy substance with sufficient hardness at room temperature.

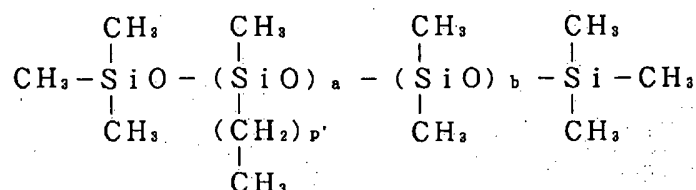
[0040] In aforementioned general formula (1), "n" is an integer from 0 to 1000, preferably, from 0 to 500, and more preferably, from 0 to 100. The organopolysiloxane
15 with "n" greater than 1000 will be too viscous in a molten state. Also, such an organopolysiloxane will have low flowability and reduced exudation from the surface of the product compounded therewith. Moreover, it will lose the mold-release property.

[0041] In order to obtain a desired chemical structure shown by general formula (1), among the groups designated by R, the amount of long-chain alkyl groups of Item (a)
20 should constitute 25 to 75%, the amount of alkoxy groups with 1 to 8 carbon atoms of Item (b) should constitute 0 to 20%, and the amount of methyl groups of item (c) should constitute 25 to 75%; "n" is within the range of 1 to 100.

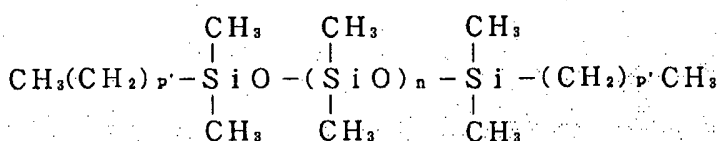
[0042] Specific structures of modified organopolysiloxanes of aforementioned general formula (1) may be those inherent in an organopolysiloxane of below-given general
25 formula (1A) with long-chain alkyl groups of Item (a) as a part of groups designated by R; an organopolysiloxane of below-given general formula (1B) with long-chain alkyl groups of Item (a) as a part of groups designated by R; an organopolysiloxane of below-given general formula (1C) with long-chain organic groups represented by the following formula: $\text{CH}_3(\text{CH}_2)_p\text{O}(\text{CH}_2)_q-$, as a part of groups designated by R; and an
30 organopolysiloxane of below-given general formula (1D) with long-chain organic groups represented by the following formula: $\text{CH}_3(\text{CH}_2)_p\text{O}(\text{CO})(\text{CH}_2)_q-$, as a part of groups designated by R.

[0043]

General formula (1A):

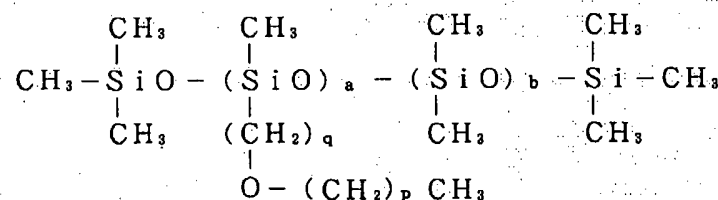


General formula (1B):

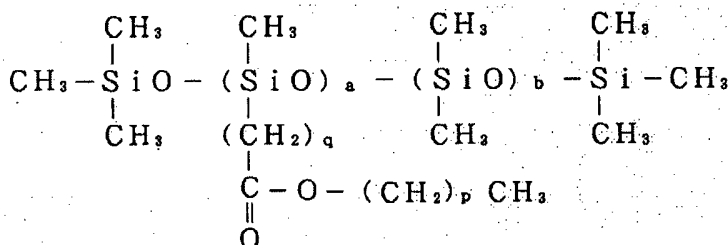


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General formula (1C):



General formula (1D):



- 10 (In the above formulae, "a" and "b" are integers with the total number of 1 to 1000; "p", "n", "p", and "q" are the same as defined above).

[Physical Characteristics of the Waxy Organopolysiloxane]

- 15 [0044] Along with the fact that the waxy organopolysiloxane of the present invention is expressed by aforementioned general formula (1), this organopolysiloxane also should satisfy the following requirements: (1) the melting point should be within the range of 55°C to 115°C; (2) viscosity at 120°C should be 50 mPa·s or below; and (3) penetration degree at 50°C should be 15 or below.

(1) Melting Point

[0045] If the melting point is below 55°C, the waxy organopolysiloxane may become liquid at room temperature, e.g., in the summer. As a result, it can melt in storage, and the product that contains this organopolysiloxane may become too soft and unsuitable for use in the waxy state. On the other hand, if the melting point exceeds 115°C, melting thereof will require an excessive amount of heat to the extent that the use of such an organopolysiloxane will become impractical. Therefore, it is recommended to have the melting point within the range of 60°C to 105°C, preferably, within the range of 70°C to 95°C. The melting point of the waxy organopolysiloxane is affected by the long-chain groups of the organopolysiloxane, in particular, the groups (a) in aforementioned general formula (1). More specifically, the waxy organopolysiloxanes may have a melting point almost the same as compounds corresponding to groups (a) (e.g., 1-eicosene in the case of eicosyl groups). Furthermore, the melting point can be raised by increasing the proportion of groups (a) among the silicon-bonded groups, or by increasing the polysiloxane chains.

(2) Viscosity at 120°C

[0046] If viscosity at 120°C exceeds 50 mPa·s, the melting time is increased, and flowability becomes insufficient for use. Therefore, it is recommended to have viscosity of 40 mPa·s or below, preferably, 25 mPa·s or below. For modified groups of the same type, the viscosity at 120°C is almost proportional to the length of the polysiloxane chain.

(3) Penetration degree at 50°C

[0047] If penetration degree at 50°C exceeds 15, the waxy organopolysiloxane becomes too soft and unsuitable for practical use. Furthermore, this makes the product compounded with the aforementioned organopolysiloxane sticky. The penetration degree should be measured in accordance with JIS K 2235. In order to obtain a waxy organopolysiloxane with penetration degree of 15 or below, it is necessary to use a compound of high hardness (e.g., with penetration degree at 50°C of 30 or below) as a precursor of groups (a).

[0048] The above condition may be fully satisfied by purifying α -olefin, or the like, as a precursor of groups (a), and the desired results can be achieved easier by providing a narrow molecular-weight distribution. The production process is disclosed, e.g., in Japanese Patent Application Laid-open No. 2003-313297, as a reprecipitation or recrystallization method.

<Method of Manufacturing the Waxy Organopolysiloxane>

[0049] The waxy organopolysiloxane of the present invention is manufactured by methods conventionally used for the preparation of modified dimethylpolysiloxanes. For example, it can be obtained by causing a hydrosilation reaction between an α -olefin and a dimethylpolysiloxane having a part of its methyl groups substituted by hydrogen atom groups.

Examples

[0050] The invention will be further described in more detail with reference to practical examples that should not be construed as limiting the scope of the invention. In the practical examples, GC, GPC, penetration degree, viscosity, and heat-absorption curves (melting point) were measured and determined as described below.

[GC]

- Equipment: [Gas chromatograph GC-390] (GL Science Co., Ltd.)
- Column: [TC-5HT] (GL Science Co., Ltd.)
- 15 - Detector: [FID]
- Measurement conditions: 70°C (10 min.) → Heating (20°C/min.) → 300°C (5 min.)

[GPC]

- 20 - Equipment: [Measurement system for Gel permeation chromatography] (Shodex Co., Inc.)
- Column: [Shodex-803L] (Showa Denko Co.)
- Detector: Refraction index (RI) detector [RI-504R] (GL Science Co.)
- Calibration curves: measurements with the use of 10 types of standard polystyrene (product of Showa Denko Co.), molecular weight: 1.2×10^3 to 2.75×10^6
- 25 - measurement conditions: at 40°C; flow of chloroform at 1.0 ml/min.; injection of 100 ml samples (concentration - 3 mass %) into the flow.

[Penetration Degree]

[0051] Penetration degree at 50°C was measured by the method specified in JIS K 2235 (1991).

30 [Viscosity (Rotary-Type Viscometer)]

- Viscometer: [Micro-viscometer LVT] (Brookfield Engineering Lab Co.)

- Cone: CP-52
- Measurement temperature: 120°C.

[Endothermic Curves - Melting Points]

[0052] Measurements were carried with a differential scanning calorimeter [DSC-6200, the product of Seiko Denshi Co., Ltd.] by using 5 mg samples. Heating-cooling cycles were carried out by first maintaining 30°C for 5 min., raising the temperature to 180°C (first heating stage) at a rate of 10°C/min., lowering the temperature to 30°C at a rate of -10°C/min. (first cooling stage), maintaining the temperature at 30°C for 5 min., and then again raising the temperature to 170°C at a rate of 10°C/min. (second heating stage). The melting point was determined as a temperature corresponding to the maximal peak on the endothermic curve of the second heating stage.

[Purification of Long-Chain α -Olefin]

[0053] 100 g of a long-chain α -olefin having 26 to 50 carbon atoms (30 carbon atoms on average) [DIALEN 30, the product of Mitsubishi Chemical Corporation, Mw = 729, Mw/Mn = 1.40] were combined with 300 g of toluene, and the α -olefin was dissolved by heating the mixture to 80°C. The solution was cooled to 60 to 70°C, and then the process was carried out by stirring in 700 g of an isopropyl alcohol (25°C) to cause the α -olefin to precipitate. The precipitate-containing solution was filtered for 2 min. in a centrifugal filter (25 cm diameter, 2000 rpm), whereby a filtration cake was obtained. The filtration cake was washed (5 min.) four times with 250 g of toluene, and then the solvent was removed by stripping the cake.

[Purification of Long-Chain (High-Grade) Alcohol]

[0054] 50 g of a long-chain alcohol having 30 to 60 carbon atoms (40 carbon atoms on average) (Unilin 550, the product of Toyo Petrolite Co.) were combined with 1050 g of a solvent mixture composed of toluene and isopropyl alcohol (respectively, 20:1 mass ratio), and the long-chain alcohol was dissolved by heating at 80°C. The solution was cooled to room temperature, and the long-chain alcohol was recrystallized. The solution with recrystallized long-chain alcohol was filtered for 30 min. in a centrifugal filter (25 cm diameter, 2000 rpm), whereby a filtration cake was obtained. The solvent was removed by stripping the cake.

[Preparation of Intermediate Starting Materials/Hydromethylpolysiloxanes (SiH1) to (SiH9)]

[0055] Two or three substances selected from hexamethyldisiloxane, tetramethyldisiloxane, octamethylcyclotetrasiloxane, and tetramethylcyclotetrasiloxane used in quantities shown in below-given Table 1 were loaded together with 30 ml of sulfuric acid into a 2-liter flask, and the flask contents were stirred for 8 hours at room temperature of 25°C. The product was neutralized and filtered to produce an intermediate starting material in the form of a hydromethylpolysiloxane (SiH1) to (SiH9) (hereinafter referred to merely as (SiH1) to (SiH9)).

[0056]

[Table 1]

Hydromethylpolysiloxane (intermediate starting material)	SiH1	SiH2	SiH3	SiH4	SiH5	SiH6
Hexamethyldisiloxane	40.3 g	51.9 g	-	49.0 g	33.3 g	3.2 g
Tetramethyldisiloxane	-	-	42.0 g	-	-	-
Octamethylcyclotetrasiloxane	-	-	58.0 g	33.1 g	30.1 g	53.5 g
Tetramethylcyclotetrasiloxane	59.7 g	48.1 g	-	17.9 g	36.6 g	43.3 g

[0057] SiH2 was subjected to heated distillation at a reduced pressure, whereby at 90°C to 120°C, 7 to 13 hPa, a distilled component (SiH7) was obtained.

[0058] SiH3 was subjected to heated distillation at a reduced pressure, whereby at 95°C to 105°C, 7 to 13 hPa, a distilled component (SiH8) was obtained.

[0059] SiH3 was subjected to heated distillation from 25°C at normal pressure to 90°C at 7 hPa, whereby a distilled component (SiH9) was obtained.

[0060] Figs. 1 to 5 show GC charts obtained by measuring aforementioned SiH1 to SiH5. Fig. 6 shows a GPC chart relating to SiH6, and Figs. 7 to 9 show GC charts relating to SiH7 to SiH9.

[Practical Example 1] (Modification with Purified Alkyl)

[0061] In accordance with proportions shown in Table 2, 12.5 g of SiH1 and 87.5 g of long-chain α -olefin [DIALEN 30] purified by the method described above were loaded into a 4-neck flask together with 100 g of xylene. A platinum catalyst (chloroplatinic acid) was added, and a hydrosilation reaction was carried out for 6 hours at 100°C.

Following this, 15 g of 1-octane [DIALEN 8] (the product of Mitsubishi Chemical Corporation) were added, and upon completion of the hydrosilation reaction, a waxy organopolysiloxane of the present invention was obtained by removing xylene at a reduced pressure.

5 [Practical Example 2] (Modification with Purified Alkyl)

[0062] A waxy organopolysiloxane of the present invention was obtained by the same method as in Practical Example 1, with the exception that, in accordance with proportions shown in Table 2, 13.0 g of SiH7 were used instead of SiH1, and the purified long-chain α -olefin [DIALEN 30] was used in the amount of 87.0 g.

10 [Practical Example 3] (Modification with Purified Alkyl)

[0063] A waxy organopolysiloxane of the present invention was obtained by the same method as in Practical Example 1, with the exception that, in accordance with proportions shown in Table 2, 16.0 g of SiH8 were used instead of SiH1, and the purified long-chain α -olefin [DIALEN 30] was used in the amount of 84.0 g.

15

[0064]

[Table 2]

			Pr. Ex. 1	Pr. Ex. 2	Pr. Ex. 3
Hydromethyl polysiloxane (SiH)	–	Type	SiH1	SiH7	SiH8
		Quantity	12.5 g	13.0 g	16.0 g
Purified long-chain α -olefin [DIALEN 30]			87.5 g	87.0 g	84.0 g

[Practical Example 4] (Modification with Purified Ester)

[0065] A 4-neck flask was filled with 25.4 g of SiH4, 16.6 g of methyl undecylenate, and 100 g of toluene. A platinum catalyst (chloroplatinic acid) was added, and a hydrosilation reaction was carried out for 6 hours at 100°C. Upon completion of the hydrosilation reaction, the reaction product was combined with 58.1 g of a long-chain alcohol (Unilin 550) purified by the above-described method and 0.5 g of titanium tetraisobutoxide, and the product was heated for 2 hours under toluene reflux conditions.

25 Following this, toluene was removed at a reduced pressure.

[Comparative Example 1] (Modification with Non-Purified Alkyl)

[0066] A 4-neck flask was filled with 21 g of SiH₆, 79 g of long-chain α -olefin [DIALEN 30] (the product of Mitsubishi Chemical Corporation), and 100 g of xylene. A platinum catalyst (chloroplatinic acid) was added, and a hydrosilation reaction was carried out for 6 hours at 100°C. Upon completion of the hydrosilation reaction, the reaction product was combined with 20 g of 1-octene [DIALEN 8] (the product of Mitsubishi Chemical Corporation), and the hydrosilation reaction was completed. The xylene was removed at a reduced pressure, and a comparative waxy organopolysiloxane was obtained.

[Comparative Example 2] (Modification with Purified Alkyl)

[0067] A 4-neck flask was filled with 23.3 g of SiH₉, 76.7 g of purified long-chain α -olefin [DIALEN 30], and 100 g of xylene. A platinum catalyst (chloroplatinic acid) was added, and a hydrosilation reaction was carried out for 6 hours at 100°C. Upon completion of the hydrosilation reaction, the reaction product was combined with 15 g of 1-octene [DIALEN 8] (the product of Mitsubishi Chemical Corporation), and the hydrosilation reaction was completed. The xylene was removed at a reduced pressure, and a comparative waxy organopolysiloxane was obtained.

[Comparative Example 3] (Modification with Purified Ester)

[0068] A 4-neck flask was filled with 14.3 g of SiH₅, 19.1 g of methyl undecylenate, and 100 g of toluene. A platinum catalyst (chloroplatinic acid) was added, and a hydrosilation reaction was carried out for 6 hours at 100°C. Upon completion of the hydrosilation reaction, the reaction product was combined with 66.7 g of a long-chain alcohol (Unilin 550) purified by the above-described method, and 0.5 g of titanium tetrakisobutoxide, and the product was subjected to 2 hour heating under toluene refluxing conditions. The toluene was removed at a reduced pressure, and a comparative waxy organopolysiloxane was obtained.

[0069] The compounds obtained in aforementioned practical and comparative examples were measured with regard to their physical properties. The results of measurements are shown below in Table 3.

[0070]

[Table 3]

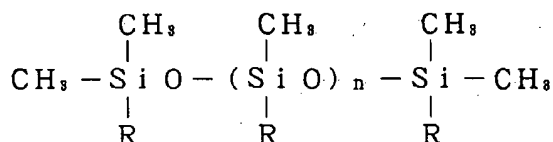
	Practical Examples				Comparative Examples		
	1	2	3	4	1	2	3
Penetration degree (at 50°C)	3	3	3	5	8	20	18
Viscosity (at 120°C) [mPa·s]	21	20	17	28	150	15	41
Melting point [°C]	75	78	78	88	72	73	89

Industrial Applicability

[0071] The waxy organopolysiloxane of the present invention that possesses the
5 above-described specific chemical structure and physical properties is suitable for use as a
wax for a thermo-sensitive transfer ribbon, thermo-sensitive paper, toners, etc.

CLAIMS

1. A waxy organopolysiloxane comprising a modified organopolysiloxane of below-given general formula (1) that has a melting point of 55°C to 115°C, a viscosity at 120°C of 50 mPa·s or below, and a penetration degree at 50°C of 15 or below:



5

[where R, independently, is:

(a) a long-chain alkyl group with 20 to 100 carbon atoms or a long-chain organic group that contains heteroatoms;

10

(b) a hydrogen atom, an alkyl group with 2 to 19 carbon atoms, an aryl group with 6 to 10 carbon atoms, an aralkyl group with 7 to 14 carbon atoms, an alkoxy group with 1 to 8 carbon atoms, or an organic group with 2 to 19 carbon atoms that contains heteroatoms; or

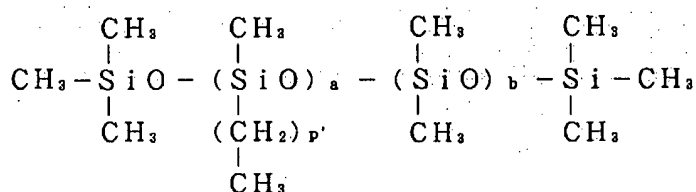
(c) a methyl group;

“n” is an integer within the range of 0 to 1000;

15

among the groups designated by R, the groups of Item (a) being contained in one molecule in the quantity of one or more.]

2. A waxy organopolysiloxane according to Claim 1, wherein the organopolysiloxane is represented by the following general formula (1A):

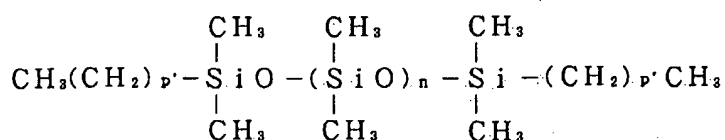


20

[where “a” and “b” are integers with the total number of 1 to 1000;

“p'” is an integer within the range of 19 to 99.]

3. A waxy organopolysiloxane according to Claim 1, wherein the organopolysiloxane is represented by the following general formula (1B):

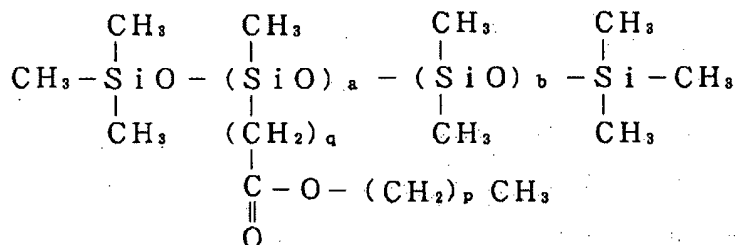


16

[where "n" is an integer within the range of 0 to 1000;

"p" is an integer within the range of 19 to 99.]

4. A waxy organopolysiloxane according to Claim 1, wherein the organopolysiloxane is represented by the following general formula (1D):



5

[where "a" and "b" are integers with the total number of 1 to 1000;

"p" is an integer within the range of 20 to 100;

"q" is an integer of 1 to 15.]

Fig. 1

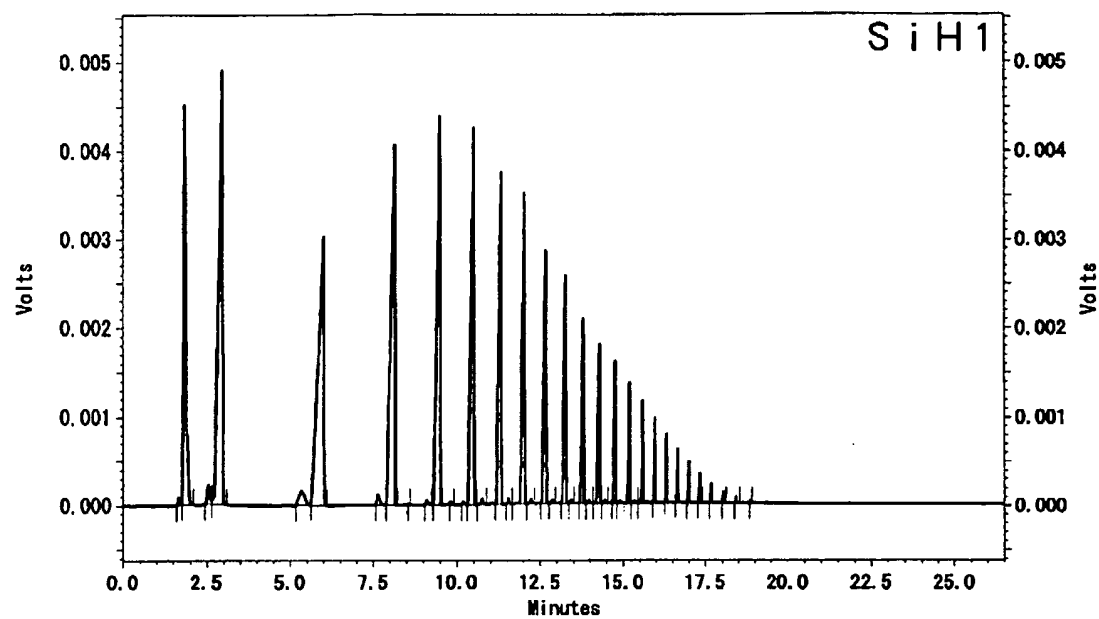


Fig. 2

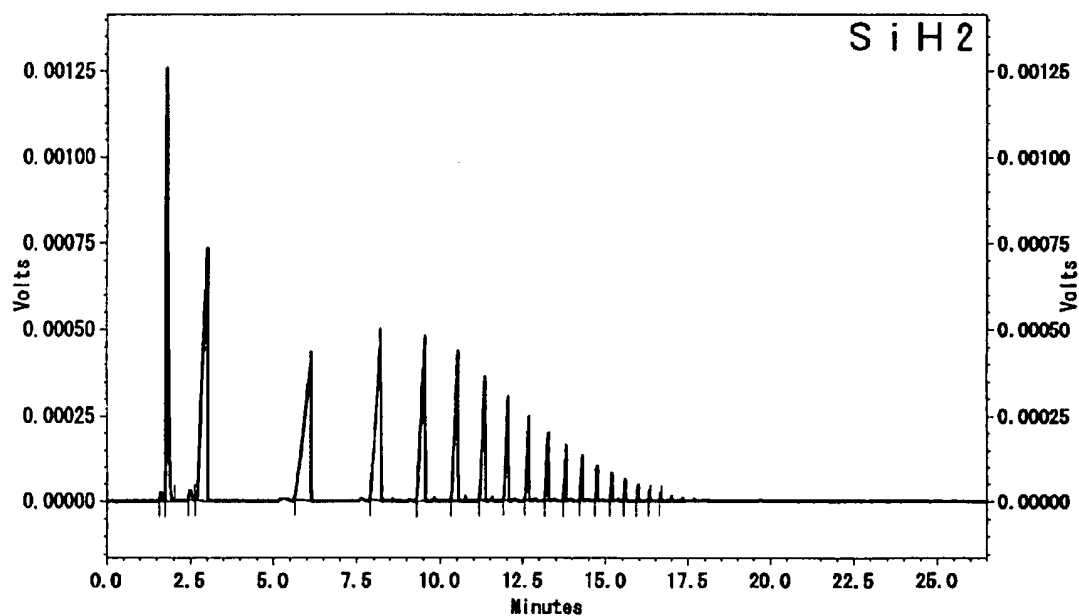


Fig. 3

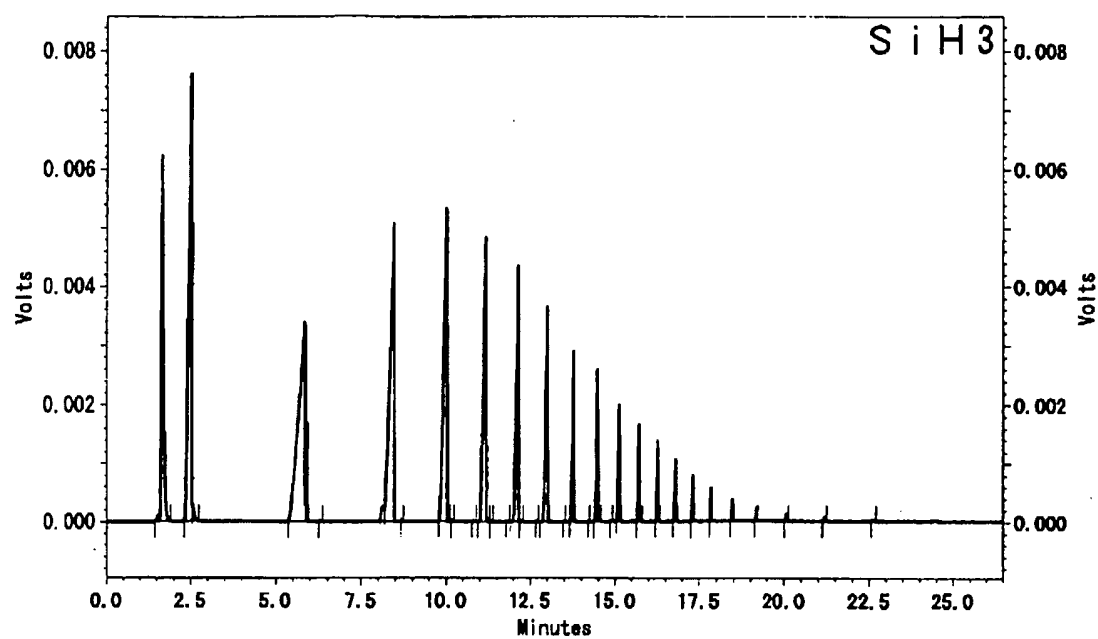


Fig. 4

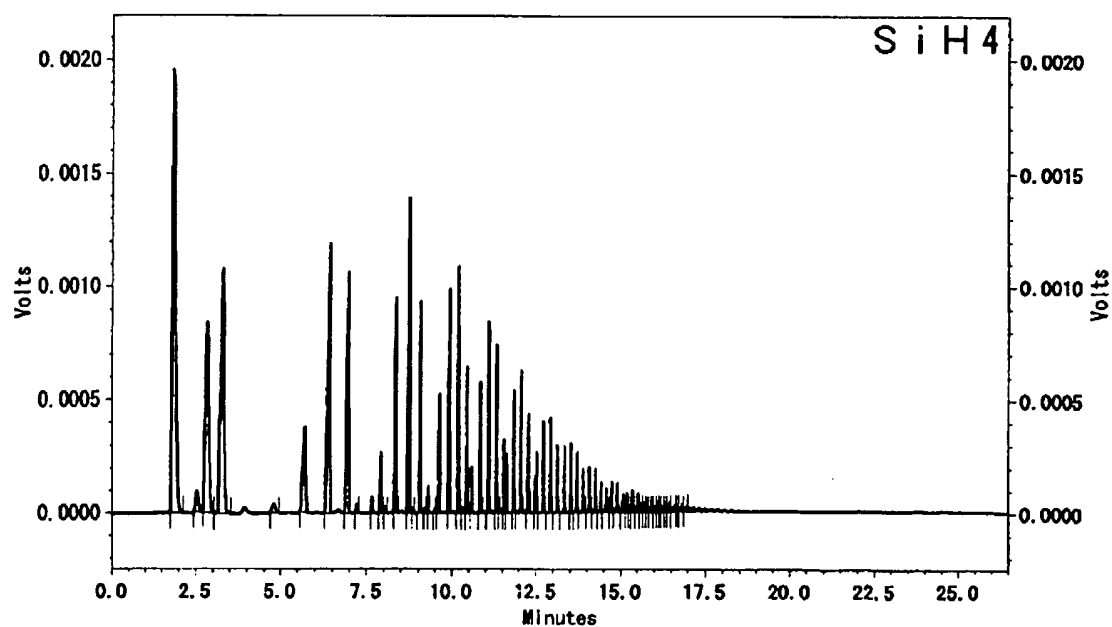


Fig. 5

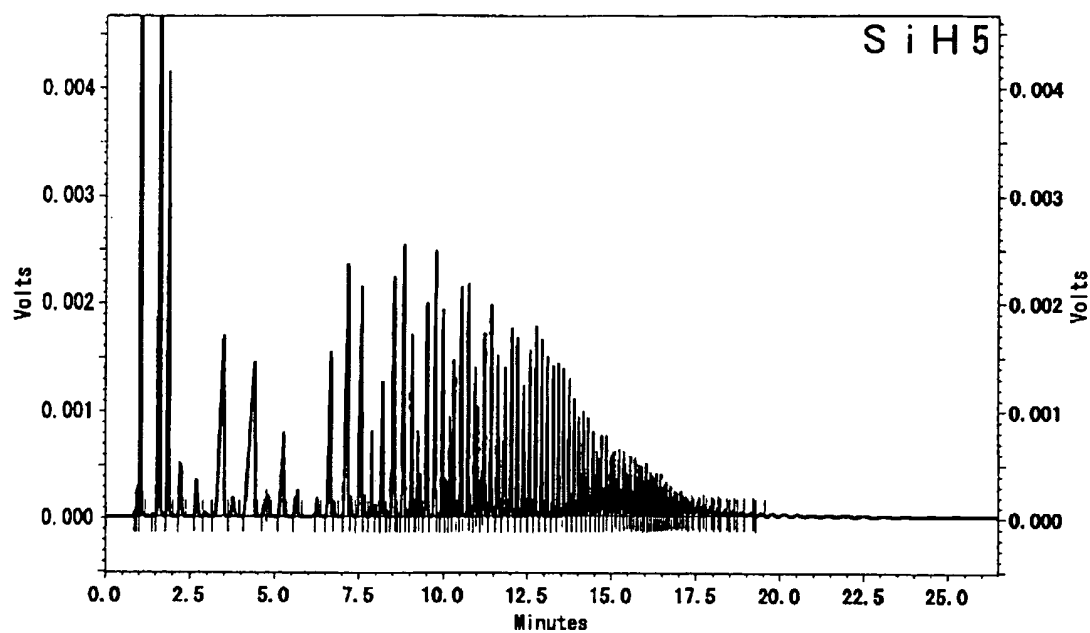


Fig. 6

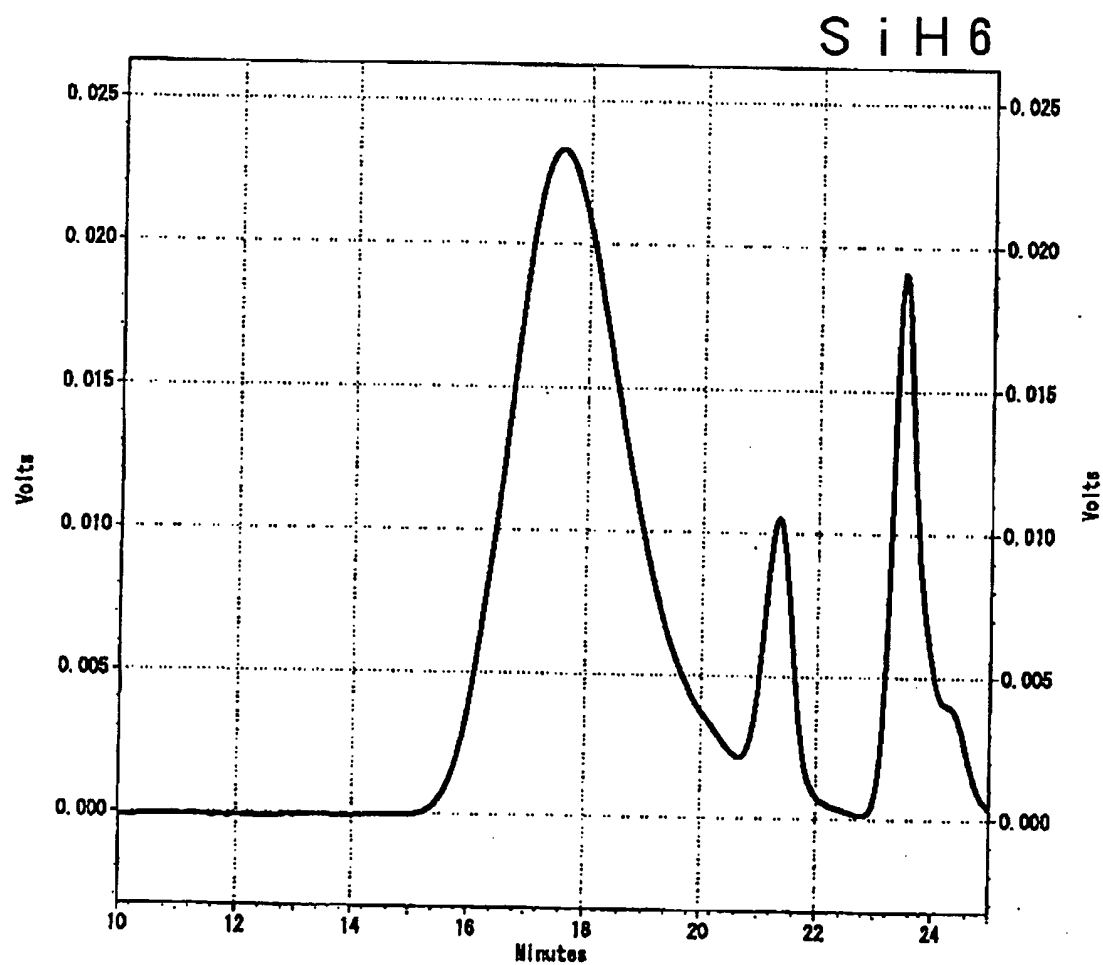


Fig. 7

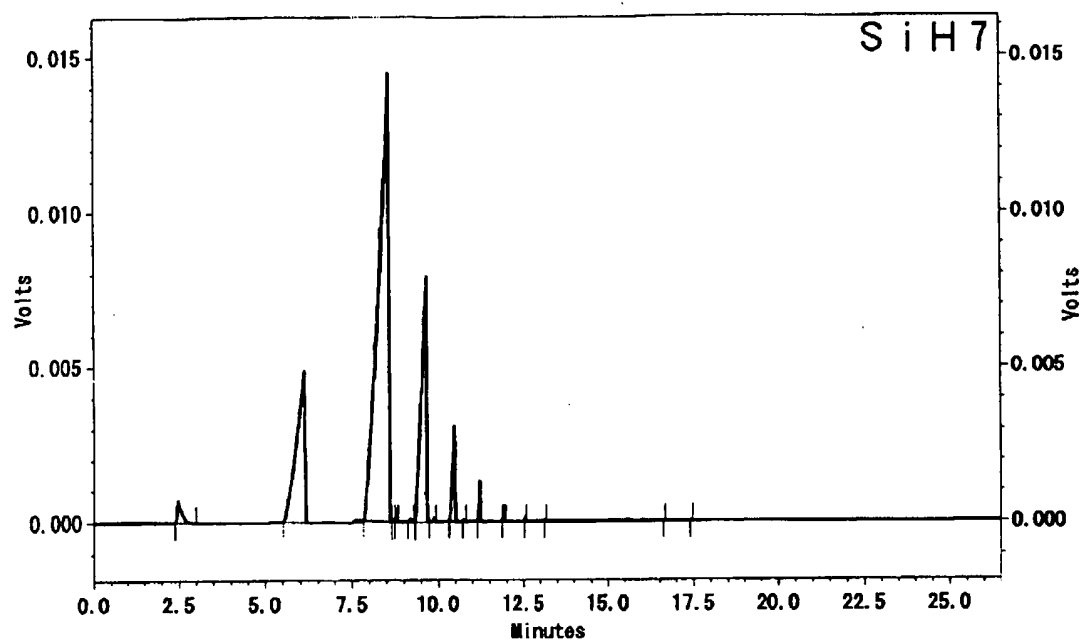


Fig. 8

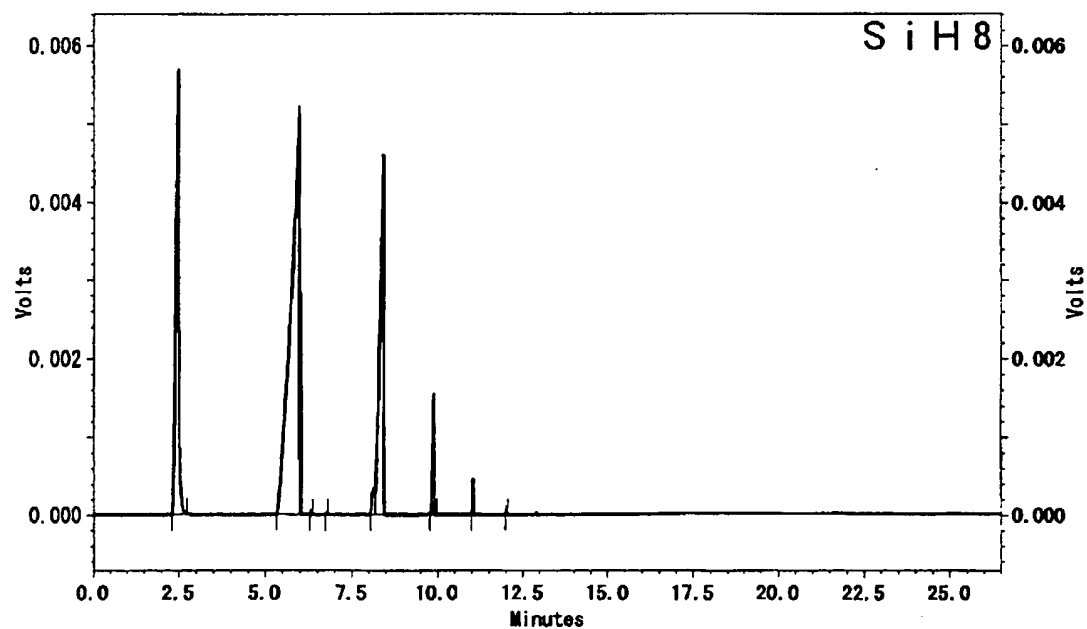
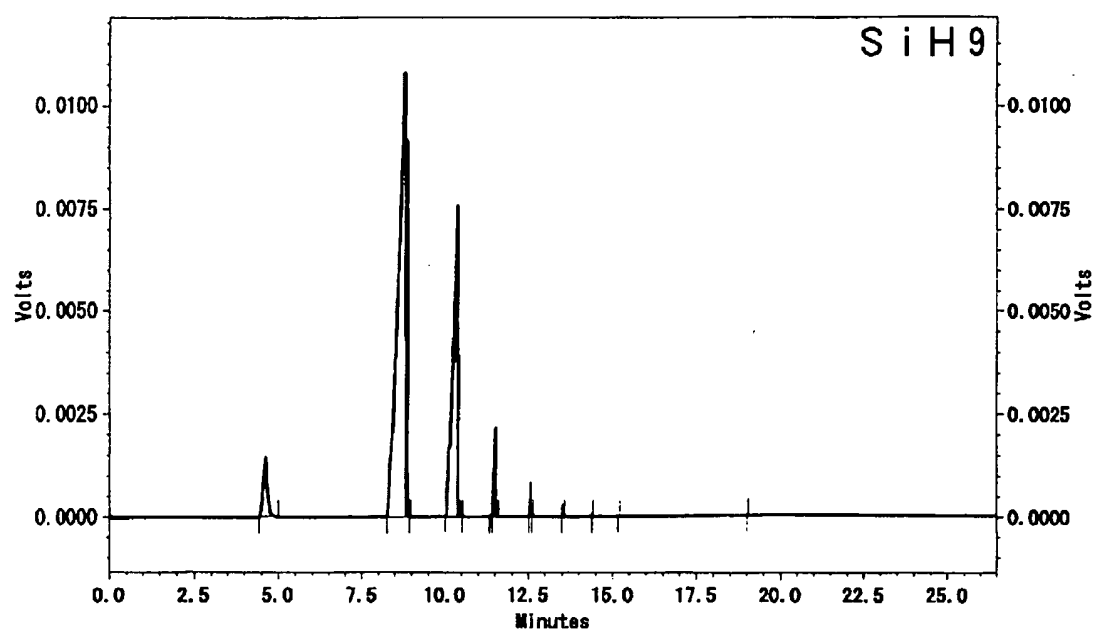


Fig. 9



INTERNATIONAL SEARCH REPORT

International Application No
PCT/JP2005/015971

A. CLASSIFICATION OF SUBJECT MATTER

C08G77/38 C08G77/442 C09G1/16

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

C08G C09G

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ, CHEM ABS Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 783 719 A (SUN ET AL)	1-3
A	21 July 1998 (1998-07-21) column 5, line 67 - column 6, line 11 claims	4
X	EP 0 878 499 A (LEXMARK INTERNATIONAL, INC) 18 November 1998 (1998-11-18)	1-3
A	claims	4

☐ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

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Date of the actual completion of the international search

7 December 2005

Date of mailing of the international search report

27/12/2005

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No
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